Fs x-ray measurement of short-range structural dynamics in VO_2 during a photo-induced phase transition.

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INTRODUCTION

The main goal of this experiment is the measurement of femtosecond structural and electronic dynamics during a phase transition in the correlated, non-magnetic oxide VO_2. This material undergoes a metal-insulator transition at 340 K, accompanied by a structural transformation between a low-T monoclinic and a high-T rutile phase. The role of electron-electron and electron-phonon correlations during this process has long been debated[1]. Current understanding favors the picture of a Mott-Hubbard insulator (low-T phase), which is turned into a simple metal upon relaxation of a structural distortion above the transition temperature.

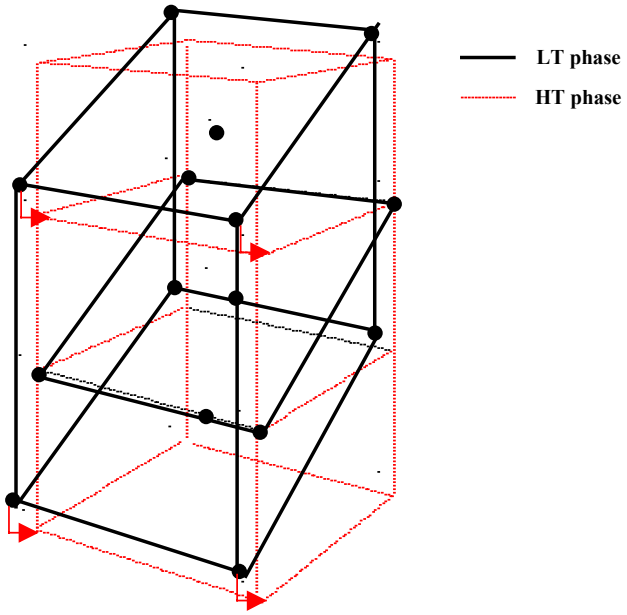

Fig. 1. Structural transition between the low-T monoclinic and high-T rutile phases of Vanadium Dioxide. A transverse optical phonon drives the transition within less than 1 picosecond.

Recent femtosecond x-ray diffraction measurements[2] have indicated that the structural transition has a very fast component, whereby long-range order between Vanadium atoms is changed within less than one picosecond. Static Raman spectra suggest that this structural transition may be impulsively induced, by photo-excitation of transverse optical phonons involving Vanadium-Vanadium distortions[3]. Further, while an additional strong V-O Raman-active mode is found (not shown in the figure) it is also known that a prominent signature of the metal insulator transition can be found near the Oxygen absorption edge[4], due to the strong hybridization of p and d orbitals from the Vanadium and Oxygen atoms, respectively.

CURRENT STATUS

The correlation between excitation of these vibrational modes and ultrafast changes in the electrical properties is the main object of our study. The experiments require combined measurement of the ultrafast electrical transition (by means of femtosecond NEXAFS spectroscopy) and of short-range structural dynamics, with sensitivity to both light (Oxygen) and heavier (Vanadium) elements (by means of femtosecond EXAFS).

The experiment will be performed using femtosecond pulses of synchrotron radiation available at the Advanced Light Source, in the spectral range between 500-800 eV, i.e. across the Vanadium L-edges and the Oxygen K-edge. The feasibility of these measurements has been demonstrated in grazing incidence geometry, where signatures of both phase transitions (electrical and structural) have been identified. For this purpose, we have performed static experiments (by varying the temperature of the sample), and time-resolved pump-probe experiments in the hundred picosecond timescale.

Because of the inconvenience of grazing incidence geometry, current efforts are dedicated to the fabrication of thin (0.05 μm), free-standing films of Vanadium Dioxide, using a combination of deposition and etching techniques. In the future, direct measurements of the EXAFS and NEXAFS component of the VO_2 transition will be conducted in a transmission geometry.

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